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The infrared spectrum of Li I

By INGMAR JOHANSSON

With 4 figures in the text

SUMMARY

Measurements have been made in the arc spectrum of lithium in the wavelength region 7000 Å–27 000 Å. The spectrum was excited in a hollow cathode and recorded photographically up to 12 000 Å with a concave grating spectrograph, and beyond 12 000 Å by means of a lead-sulfide photoconductive cell in a Pfund type scanning spectrograph. On the basis of the new measurements in combination with interferometer measurements by Meissner, Mundie and Stelson (1948) the term system has been revised and accurate formulae have been derived for the various term series.

Introduction

A relatively complete description of the arc spectrum of lithium was given as early as 1922 in the well-known monographs on series in line spectra by Paschen and Götze and by Fowler. However, despite subsequent work on this spectrum, accurate term values for the ^2F series and the higher ^2P terms have been lacking. The present contribution consists of an investigation of the infrared region of the lithium spectrum by means of a hollow-cathode discharge and sensitized photographic plates as well as a lead-sulfide cell, which has yielded accurate values of the ^2P and ^2F terms. The new values for the ^2F terms provide a good test of the polarization formula for non-penetrating orbits.

The light source

Since the lithium spectrum is very sensitive to Doppler broadening and Stark effect, a hollow cathode, giving moderate temperatures and low field strengths, was chosen as a light source. The discharge tube was a copy of that used by Risberg (1956) for his study of the sodium and potassium spectra. A drawing showing this light source is given in Risberg's paper. The cathode consisted of a hollow iron cylinder having a length of 100 mm, an inner diameter of 15 mm and a wall thickness of 3 mm. It was closed in one end, and the opening at the other end was constricted to 9 mm. The cylinder was cooled at the closed end with running water. In order to maintain a uniform temperature along the cathode the wall of the cylinder was made rather thick. A copper cathode would have been preferable for that purpose, but, unfortunately, molten lithium metal forms an alloy with copper. Lithium metal was introduced into the cathode in small pieces from the front after removing a quartz window which was attached to the anode by means of vacuum grease. The metal used was relatively impure. At first the spectrum was a nearly pure sodium spectrum because of the lower boiling point of this metal. By maintaining the discharge and slowly raising the temperature the sodium could be so completely removed by distillation that the yellow resonance doublet became only just discernible. With a suitable temperature in the cathode an intense lithium spectrum was then obtained. The discharge was run in an atmosphere of neon or argon at a pressure of 0.2–1 mm Hg. The temperature of the cathode was adjusted by changing the electric current and the flow of water in the cooling system. The voltage between the anode and the cathode was about 200 V, and the current was generally kept between 0.6 and 0.8 A. When the temperature was too high the metal distilled out rapidly, and when it was too low the metal spectrum faded and gave place to a very pure, intense rare-gas spectrum. On account of the getter effect of the lithium vapour this discharge could be run for several days without using circulation or charcoal traps. The purifying effect of alkali metals on hollow-cathode discharges was mentioned by Schüler in 1926.

The observations

For the observations in the photographic region a 21-foot concave grating in a Wadsworth mounting was used. The grating, having 400 lines/mm and a total of 60 000 lines, gave a dispersion of 7.5 Å/mm in the first order. Eastman Kodak spectrosopic plates of types *Z*, *Q*, *M* and *N* were used. They were all hypersensitized before exposure. The exposures were some times extended to ten hours in order to bring out the faintest lines. Only first order lines were measured. Neon, argon, helium and xenon provided reference lines. The neon and argon lines were mainly adopted standards; the remaining wavelengths were based on interferometric measurements or calculated from accurately known levels. The lithium lines 9955 Å and 10 032 Å were measured on two plates only, all other lines on three or more plates. The measured lines are collected in Table 1. A few more lines were observed, but they were too faint and diffuse to permit accurate measurement.

In the lead-sulfide region the lithium spectrum was recorded with a Pfund-type scanning spectrograph. Two different plane replica gratings from Bausch and Lomb were used. One, with a blaze at 20 000 Å, has 300 grooves/mm over a ruled area of 127 × 153 mm, and the other, with a blaze at 16 000 Å, has 600 grooves/mm over a ruled area of 102 × 128 mm. As the focal length of the concave mirrors is only one metre, the apertures amount to about $f/5$ and $f/6$ respectively. Owing to these great apertures, the theoretical resolving power of the spectrograph would be reached only by using very narrow slits. The actual resolution, however, was limited by the noise of the photocell and the amplifier, because the slits had usually to be widened in order to get a strong enough signal. Wavelengths were determined by means of a wave-number scale which was established with the aid of a Fabry–Perot interferometer. Several such methods have been worked out elsewhere, the present one being similar to that described by Eshbach and Fisher (1954). The scale was calibrated by using adopted rare-gas standards in the second or third order. For the lithium line 13 557 Å use was made of first order argon lines from interferometric measurements by Humphreys and Paul (1957). The agreement of individual measurements as well as the combination principle test indicate an accuracy in the measurements of

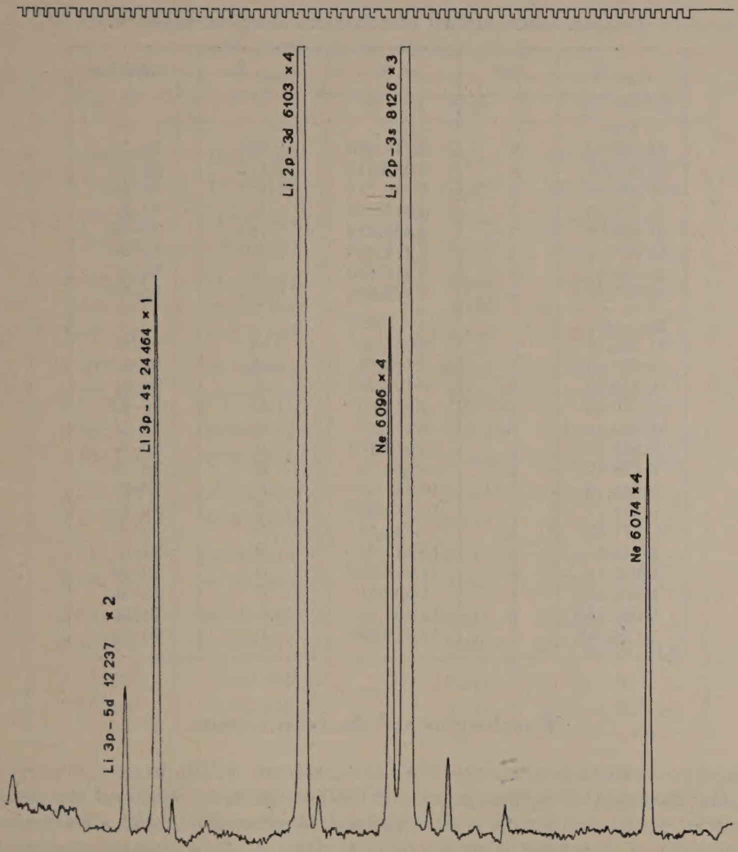

Fig. 1. A record from the lead-sulfide spectrograph showing four lithium lines, all of different orders, together with two fourth-order standard lines of neon. The wave-number scale is seen at the top. This is a survey record; in measurement records the dispersion was about ten times larger.

±0.02 Å. Fig. 1 is a record showing standard lines and lithium lines of different orders as well as the wave-number scale.

In the region 1–2 μ the water vapour has two strong absorption bands,[1] the very strongest line of which falls at 18 705 Å. This line may cause an asymmetric absorption of the lithium line at 18 703 Å, resulting in a shift in the observed wavelength. To prevent such errors the whole spectrograph was enclosed in an envelope of polyethylene, which made it possible to dry the air inside. After drying the observed wavelength of λ 18 703 increased by 0.12 Å, as compared to the result obtained in moist air. Some other lines which could be expected to be similarly influenced by water vapour absorption were also remeasured with dry air in the spectrograph.

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[1] See for instance Nelson (1949).

Table 1. Lines of Li I observed in the present work.

λ_{air}, Å	Int	σ, K	σ_{calc}, K	Transition
PbS				
26 877.82	8	3 719.525	9.532	$3s$–$3p$
24 464.66	6	4 086.415	6.412	$3p$–$4s$
19 274.78	4	5 186.710	P	$3d$–$4p$
18 703.09	7	5 345.251	F	$3d$–$4f$
17 546.05	7	5 697.733	7.731	$3p$–$4d$
13 557.75	4	7 373.838	3.841	$3p$–$5s$
12 793.31	5	7 814.450	F	$3d$–$5f$
12 237.67	4	8 169.255	9.270	$3p$–$5d$
Photogr.				
11 032.09	1	9 061.99	1.98	$3p$–$6s$
10 976.06	0	9 108.24	8.23	$3d$–$6p$
10 919.07	3*d*	9 155.78	F	$3d$–$6f$
10 510.60	3*d*	9 511.60	1.65	$3p$–$6d$
10 032.81	2*d*	9 964.57	F	$3d$–$7f$
9 955.09	2	10 042.36	S	$3p$–$7s$
9 686.37	2	10 320.95	D	$3p$–$7d$
9 530.73	1*d*	10 489.50	F	$3d$–$8f$
9 376.71	1	10 661.80	S	$3p$–$8s$
9 217.32	2*d*	10 846.16	D	$3p$–$8d$
9 214.61	1*d*	10 849.35	F	$3d$–$9f$
8 921.14	0	11 206.26	D	$3p$–$9d$
8 465.352	4	11 809.61	P	$3s$–$5p$
7 582.169	3	13 185.21	5.21	$3s$–$6p$
7 135.040	1	14 011.48	P	$3s$–$7p$

Wavelengths and the term system

The most accurate measurements in the arc spectrum of lithium are due to Meissner, Mundie and Stelson (1948), who measured the resonance doublet and the sharp and diffuse series up to $n = 6$ with a Fabry–Perot interferometer and an atomic-beam light source. Their wavelengths are accurate to 0.001 Å. They give also the fine structure of $nd\,^2D$ and $2p\,^2P$. Measurements had previously been made by Werner (1927) in the same series up to 11*s* and 12*d*. France (1930) observed the principal series in absorption up to $n = 32$ with a grating spectrograph, and Lidén and Starfelt (1952) measured the same series up to 5*p* with a Fabry–Perot interferometer and to 13*p* with a quartz spectrograph. In addition, Datta and Bose (1935) have measured the lowest members of the principal as well as the sharp and diffuse series. Their results, and those of some other authors, are quoted and discussed in the paper by Meissner *et al.*

The light source used in the present work gives a rather undisturbed radiation, which is essential for an accurate description especially of the fundamental series. Furthermore, since the measurements concern transitions between terms with quantum numbers $n = 3$ and higher, which give relatively long wave-lengths, a wave-number accuracy is obtained which is considerably better than that of previous measurements, except those of Meissner *et al.*

The new wave-lengths are collected in Table 1 which contains twenty-three lines from 26 878 Å to 7135 Å. Those falling in the photographic region are all faint lines

Table 2. The interferometric measurements by Meissner, Mundie and Stelson (1948), with recalculated wave-numbers.

λ_{air}, Å	σ, K	$\Delta\sigma$	σ_{calc}	Combination
8 126.452	12 302.111	0.334	2.110	$2p\,^2P_{3/2}$–$3s\,^2S_{1/2}$
8 126.231	12 302.445		2.446	$2p\,^2P_{1/2}$–$3s\,^2S_{1/2}$
6 707.912	14 903.654	0.336	3.654	$2s\,^2S_{1/2}$–$2p\,^2P_{1/2}$
6 707.761	14 903.990		3.990	$2s\,^2S_{1/2}$–$2p\,^2P_{3/2}$
6 103.664	16 379.067	0.041	9.069	$2p\,^2P_{3/2}$–$3d\,^2D_{3/2}$
6 103.649	16 379.108	0.297	9.108	$2p\,^2P_{3/2}$–$3d\,^2D_{5/2}$
6 103.538	16 379.405		9.405	$2p\,^2P_{1/2}$–$3d\,^2D_{3/2}$
4 971.745	20 108.052	0.339	8.054	$2p\,^2P_{3/2}$–$4s\,^2S_{1/2}$
4 971.661	20 108.391		8.390	$2p\,^2P_{1/2}$–$4s\,^2S_{1/2}$
4 602.894	21 719.379	0.321	9.379	$2p\,^2P_{3/2}$–$4d\,^2D_{5/2}$
4 602.826	21 719.700		9.700	$2p\,^2P_{1/2}$–$4d\,^2D_{3/2}$
4 273.127	23 395.484	0.334	5.483	$2p\,^2P_{3/2}$–$5s\,^2S_{1/2}$
4 273.066	23 395.818		5.819	$2p\,^2P_{1/2}$–$5s\,^2S_{1/2}$
4 132.618	24 190.915	0.328	0.915	$2p\,^2P_{3/2}$–$5d\,^2D_{5/2}$
4 132.562	24 191.243		1.243	$2p\,^2P_{1/2}$–$5d\,^2D_{3/2}$
3 985.538	25 083.622	0.333	3.621	$2p\,^2P_{3/2}$–$6s\,^2S_{1/2}$
3 985.485	25 083.955		3.957	$2p\,^2P_{1/2}$–$6s\,^2S_{1/2}$
3 915.346	25 533.294	0.332	3.294	$2p\,^2P_{3/2}$–$6d\,^2D_{5/2}$
3 915.295	25 533.626		3.626	$2p\,^2P_{1/2}$–$6d\,^2D_{3/2}$

which have not been observed before. The remaining eight lines have been measured by Paschen (1908, 1910). His wave-lengths differ from the present values by amounts ranging from −11 to +15 Å.

Two expected transitions have not been found. One is $3d$–$5p$ with a predicted wave-length of 12 929 Å, which falls near the sensitivity limit of photographic emulsions. Since the sensitivity of the lead-sulfide cell, as well as the blaze of the gratings, are less favourable here than at longer wavelengths, the line could not be detected photoelectrically either. The other missing transition is $3s$–$4p$, expected at 10 791 Å. Despite the fact that the line falls in a region where the Z-emulsion has its maximum sensitivity, it was impossible to detect it. This is obviously a case of anomalous intensity.

The calculation of the term system started from the interferometer measurements of Meissner, Mundie and Stelson, which establish the connection of the low terms $2s^2S$ and $2p^2P$ with ns^2S and nd^2D up to $n=6$ (see Fig. 2). The results given by these authors are reproduced in Table 2 with the wave-numbers corrected according to Edlén's (1953) air-dispersion formula. The relative term values obtained from the data of Table 2 should be correct to ± 3 mK (1 mK $= 0.001$ cm^{-1}). In order to combine them with the measurements of Table 1, with unresolved fine-structures, the centre of gravity of the fine-structure levels was calculated for each term. As

Table 3. Observed term values of Li I.

T and n^* refer to the centre of gravity of the fine-structure components.

Term symbol	E cm^{-1}	ΔE	T cm^{-1}	n^*
$2s\,^2S$	0.000		43 487.150	1.588 471 9
$3s\,^2S$	27 206.100		16 281.050	2.596 084 3
$4s\,^2S$	35 012.044		8 475.106	3.598 217 6
$5s\,^2S$	38 299.473		5 187.677	4.599 107
$6s\,^2S$	39 987.611		3 499.539	5.599 569
$7s\,^2S$	40 967.99		2 519.16	6.599 82
$8s\,^2S$	41 587.43		1 899.72	7.600 03
$2p\,^2P_{\frac{1}{2}}$	14 903.654	0.336	28 583.272	1.959 314 0
$^2P_{1\frac{1}{2}}$	14 903.990			
$3p\,^2P$	30 925.632		12 561.518	2.955 554 3
$4p\,^2P$	36 469.79		7 017.36	3.954 332
$5p\,^2P$	39 015.71		4 471.44	4.953 78
$6p\,^2P$	40 391.31		3 095.84	5.953 48
$7p\,^2P$	41 217.58		2 269.57	6.953 25
$3d\,^2D_{1\frac{1}{2}}$	31 283.059	0.039	12 204.068	2.998 525 1
$^2D_{2\frac{1}{2}}$	31 283.098			
$4d\,^2D_{1\frac{1}{2}}$	36 623.354	0.015	6 863.787	3.998 325 1
$^2D_{2\frac{1}{2}}$	36 623.369			
$5d\,^2D_{1\frac{1}{2}}$	39 094.897	0.008	4 392.248	4.998 234
$^2D_{2\frac{1}{2}}$	39 094.905			
$6d\,^2D_{1\frac{1}{2}}$	40 437.280	0.004	3 049.868	5.998 180
$^2D_{2\frac{1}{2}}$	40 437.284			
$7d\,^2D$	41 246.59		2 240.56	6.998 12
$8d\,^2D$	41 771.79		1 715.36	7.998 03
$9d\,^2D$	42 131.89		1 355.26	8.998 06
$4f\,^2F$	36 628.33		6 858.82	3.999 774
$5f\,^2F$	39 097.53		4 389.62	4.999 732
$6f\,^2F$	40 438.86		3 048.29	5.999 73
$(7f\,^2F)$[a]	41 247.65		2 239.50	6.999 78
$(8f\,^2F)$[a]	41 772.58		1 714.57	7.999 86
$(9f\,^2F)$[a]	42 132.43		1 354.72	8.999 85

[a] These values probably include part of $ng\,^2G$, $nh\,^2H$ etc.

the next step the term $3p\,^2P$ was derived from seven different combinations (cf. Fig. 2). The agreement of individual values suggests an accuracy of about ±5 mK in the position of $3p\,^2P$ thus obtained. All remaining terms were then determined relative to $3s\,^2S$, $3p\,^2P$ or $3d\,^2D$. The term values are collected in Table 3, and a Grotrian diagram of the term system is shown in Fig. 2.

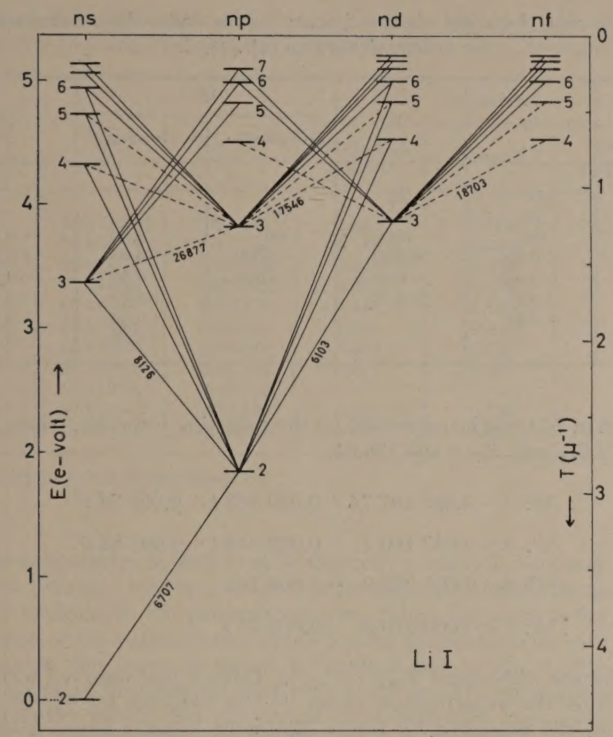

Fig. 2. Grotrian diagram of Li I. Solid lines indicate transitions observed photographically, while dashed lines represent lead-sulfide observations. All combinations with $2p$ shown in the diagram have been measured by Meissner, Mundie and Stelson. Only those transitions that were used in calculating the term system are included. Hence the higher members of the principal series are not shown.

Determination of the series limit and series formulae

For the purpose of calculating the series limit one can choose between the ^2S and the ^2D series. The ^2D series is preferable for several reasons. Meissner considers this series more reliable because of the greater intensity of the lines. Moreover, the ^2D series is so hydrogen-like that it can be assumed to follow exactly a simple, two-constant Ritz formula. The limit and the constants of the Ritz formula were determined from the four lowest ^2D terms in such a way that $3d$ and $4d$ are exactly reproduced by the formula, and the deviations between observed and calculated values of $5d$ and $6d$ are equal with opposite signs. For the limit was obtained the value 43 487.150 cm^{-1} which is thought to be correct within 5 mK. The limit being fixed it was then possible to work out Ritz formulae for the ^2S and ^2P series. Here it proved necessary to use extended Ritz formulae with three constants.[1] As a result, the four

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[1] See, for instance, Edlén and Risberg (1956).

Table 4. Differences between observed term values and values calculated from the Ritz formulae (1)–(4).

n	^2S	^2P	^2D Interferom.	^2D Grating	^2F
2	(0)	(0)			
3	(0)	(0)	(0)		
4	(0)	(0)	(0)	0.00	−0.02
5	+0.005	+0.01	−0.002	+0.01	+0.02
6	−0.004	+0.01	+0.002	+0.05	−0.01
7	+0.01	+0.04		+0.02	−0.04
8	−0.01			+0.05	−0.06
9				+0.02	−0.05

series observed in Li I can be expressed by the following formulae, where $\delta = n - n^*$, $t = (n^*)^{-2} = T / R_{\mathrm{Li}}$ and $R_{\mathrm{Li}} = 109\,728.64$:

$$^2S:\ \delta = 0.399\,491\,74 + 0.029\,483\,t + 0.002\,24\,t^2 \tag{1}$$

$$^2P:\ \delta = 0.047\,160\,1 - 0.022\,816\,t - 0.007\,82\,t^2 \tag{2}$$

$$^2D:\ \delta = 0.001\,932\,0 - 0.004\,11\,t \tag{3}$$

$$^2F:\ \delta = 0.000\,310\,3 - 0.001\,27\,t \tag{4}$$

As shown by the differences $T_{\mathrm{obs}} - T_{\mathrm{calc}}$ in Table 4 the observed term values are reproduced within the experimental errors by the formulae for ^2S, ^2P and ^2D. On the other hand, the measured ^2F terms cannot be exactly represented by a Ritz formula, as is seen from the diagram in Fig. 3 and from the values of n^* in Table 3.

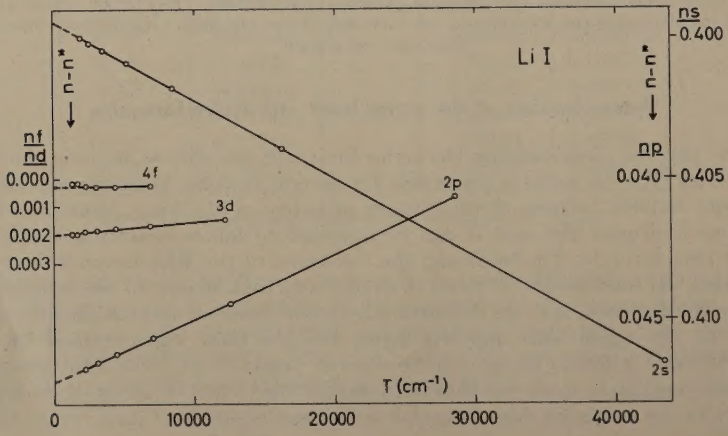

Fig. 3. Series diagram showing $n - n^*$ as a function of absolute term value.

Table 5. Terms and wavelengths of the principal series for $n = 3 - 13$; the term values for $8 - 13\,p$ are extrapolated by means of the series formula (2).

Term	E_{np}	λ_{air}, calc	λ observed		
			Lidén and Starfelt	France	Datta and Bose
$3p$	30 925.632	3232.631	2.634[a]	2.660	2.623
$4p$	36 469.79	2741.184	1.186[a]	1.204	1.188
$5p$	39 015.71	2562.302	2.305[a]	2.312	
$6p$	40 391.31	2475.032	5.057	5.061	
$7p$	40 217.58	2425.413	5.414	5.426	
$8p$	41 752.42	2394.342	4.355	4.386	
$9p$	42 118.25	2373.543	3.548	3.541	
$10p$	42 379.49	2358.911	8.917	8.928	
$11p$	42 572.51	2348.215	—	—	
$12p$	42 719.14	2340.154	0.161	0.153	
$13p$	42 833.14	2333.925	3.942	3.936	

[a] Interferometric determinations.

The probable explanation is that even in the hollow-cathode discharge the ^2F terms are somewhat disturbed by Stark effect. The formula (4), which is shown as a straight line in Fig. 3, is fitted to the undisturbed term values discussed below.

A comparison of the extrapolated values for the principal series with earlier direct measurements in this series is shown in Table 5 and in Fig. 4. The wavelengths measured in emission with interferometer by Lidén and Starfelt agree within 0.003 Å with the calculated values. For the higher series members, measured in absorption, the discrepancies plotted in Fig. 4 are seen to be considerable; in some cases the observed values fall half-way between two calculated positions.

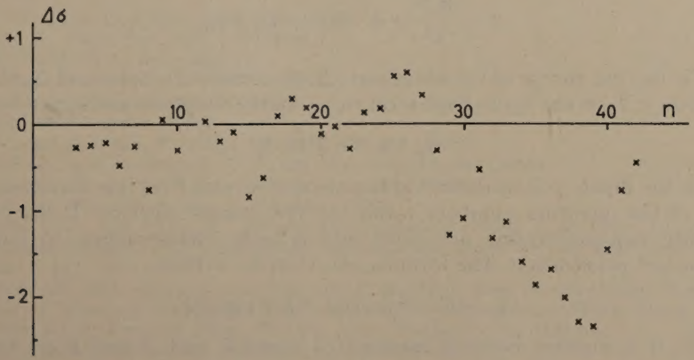

Fig. 4. Differences $\Delta\sigma$ between observed and calculated wave numbers for the principal series of Li I, plotted against the quantum number n. The observed values are from France ($n = 3 - 32$) and from Bevan ($n = 33 - 42$), while the calculated values are obtained from the series formula (2).

Table 6. Term values calculated with the polarization formula (7).

$\Delta_p = 0.190\,73\ P(1+0.188\ r)$; $T_{\text{calc}} = T_{\text{H}} + \Delta_p$.

	T_{H}	$(\Delta_p)_{\text{calc}}$	T_{calc}	T_{obs}
3*d*	12 192.104	11.964	12 204.068	12 204.068
4*d*	6 858.059	5.728	6 863.787	6 863.787
5*d*	4 389.157	3.095	4 392.252	4 392.248
6*d*	3 048.025	1.842	3 049.867	3 049.868
7*d*	2 239.365	1.179	2 240.544	2 240.56
8*d*	1 714.514	0.799	1 715.312	1 715.36
9*d*	1 354.677	0.565	1 355.242	1 355.26
10*d*	1 097.288	0.414	1 097.702	
4*f*	6 858.049	0.782	6 858.83	6 858.82
5*f*	4 389.152	0.449	4 389.60	4 389.62
6*f*	3 048.022	0.275	3 048.30	3 048.29
7*f*	2 239.363	0.179	2 239.54	2 239.50
8*f*	1 714.512	0.122	1 714.63	1 714.57
9*f*	1 354.677	0.087	1 354.76	1 354.72
10*f*	1 097.287	0.064	1 097.35	
5*g*	4 389.149	0.107	4 389.26	
6*g*	3 048.021	0.068	3 048.09	
7*g*	2 239.362	0.046	2 239.41	
8*g*	1 714.511	0.032	1 714.54	
9*g*	1 354.676	0.023	1 354.70	
10*g*	1 097.287	0.017	1 097.30	

The polarization formula

In order to calculate undisturbed values of the ^2F terms one can apply the polarization theory.[1] According to this theory the quantum defect for non-penetrating orbits is mainly an effect of polarization of the atom core. Following a treatment by Edlén (1958) the term values of non-penetrating orbits can be written

$$T = \frac{R\zeta^2}{n^2} + \Delta_r + \Delta_p = T_{\text{H}} + \Delta_p, \tag{5}$$

where ζ is the net charge of the atom core, Δ_r the relativistic term and Δ_p the polarization energy. T_{H} is the hydrogenic term value. In the simplest case Δ_p can be written:

$$\Delta_p = \alpha\zeta^4 \times P(n, l), \tag{6}$$

where α is the dipole polarizability[2] of the atom core, and $P(n, l)$ is a known, rational function of the quantum numbers n and l of the valence electron. If the orbits are not strictly non-penetrating one must add a term corresponding to quadrupole distortion and penetration. The formula can then be written:

$$\Delta_p = A \times P(n, l)[1 + k \times r(n, l)], \tag{7}$$

where $r(n, l)$ is another rational function of n and l, and A and k are treated as adjustable parameters. For Li I the parameter A is simply the dipole polarizability α.

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[1] See, for example, Bockasten (1956) I and II.

[2] Expressed in units of $a_0^3 = 1.4816 \times 10^{-25}$ cm^3.

Bockasten (1956, II) has mentioned that the ^2D series of Li I can be represented by this two-constant polarization formula with the same accuracy as by a Ritz formula. In the same paper he gives tables of $\varphi(n,l)=P(n,l)/R$ and $\psi(n,l)=r(n,l)\times\varphi(n,l)$.

From the two lowest members of the ^2D series in Li I one obtains the values $A=0.19073$ and $k=0.188$. The polarization formula can then be used to calculate undisturbed values of ^2F and ^2G terms. The Ritz formula (4), where the coefficients have been determined from the undisturbed values of the two lowest ^2F terms, gives the same extrapolated term values as the polarization formula. Table 6 gives Δ_p and calculated term values for the ^2D, ^2F and ^2G series. A comparison with the observed values reveals small but not quite insignificant differences. It is most plausible to explain them as due to Stark effect, especially in view of the noticeable broadening observed for the corresponding lines. A Stark effect would increase the T values of the higher ^2D terms and of the ^2F terms, except $4f\,^2F$, which should be decreased. This is in agreement with the sign of the differences in Table 4 for the ^2D and the two lowest ^2F terms but not for the higher ^2F terms. The explanation of the latter anomaly is probably that for the higher members of the fundamental series the measured lines correspond to an unresolved blend (separation 0.2 cm^{-1} or less) of $3d-nf$ with $3d-ng$, the latter transitions being induced by Stark effect.

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Department of Physics, University of Lund, Lund, Sweden, November 1958.

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